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Plutonium measurements by accelerator mass spectrometry at LLNL

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Abstract

Mass spectrometric methods provide sensitive, routine, and cost-effective analyses of long-lived radionuclides. Here we report on the status of work at Lawrence Livermore National Laboratory (LLNL) to develop a capability for actinide measurements by accelerator mass spectrometry (AMS) to take advantage of the high potential of AMS for rejection of interferences. This work demonstrates that the LLNL AMS spectrometer is well-suited for providing high sensitivity, robust, high throughput measurements of plutonium concentrations and isotope ratios. Present backgrounds are $\sim 2 \times 10^7$ atoms per sample for environmental samples prepared using standard alpha spectrometry protocols. Recent measurements of $^{239+240}\text{Pu}$ and ^{241}Pu activities and $^{240}\text{Pu}/^{239}\text{Pu}$ isotope ratios in IAEA reference materials agree well with IAEA reference values and with alpha spectrometry and recently published ICP-MS results. Ongoing upgrades of the AMS spectrometer are expected to reduce backgrounds below 1×10^6 atoms per sample while allowing simplifications of the sample preparation chemistry. These simplifications will lead to lower per-sample costs, higher throughput, faster turn around and, ultimately, to larger and more robust data sets.

1. Introduction

Plutonium and other actinides are distributed throughout the environment as a result of nuclear weapons testing, fuel reprocessing, reactor operations and, to a lesser extent, accidental releases.^{1,2} Plutonium is found in fresh and sea waters,^{3,4} sediments,^{5,6} aerosol particles, ice cores, and groundwaters.⁷ Because of the prevalence and potential health effects of these radionuclides, effort is devoted worldwide to actinide studies including basic actinide chemistry, environmental concentrations, speciation and partitioning, transport, evaluations of exposure pathways,^{1,8,9} toxicokinetic studies,¹⁰ bioassays of exposed populations,¹¹⁻¹⁵ risk assessments,¹⁶ and nuclear non-proliferation and national security.¹⁷ Effects such as “hot” particles,¹⁸ resuspension,¹⁶ bioconcentration,¹⁹ and colloidal transport⁷ can complicate the transport, exposure pathways, and subsequent bioavailability. Daughters of pre-anthropogenic plutonium have also been studied for geochronology.²⁰

Measurements provide the foundation for these activities, and the most commonly applied analytical tool is alpha spectrometry. Many applications, particularly those most relevant to human health effects, require significantly higher sensitivity than is available with alpha spectrometry ($\sim 10^8$ atoms $^{239,240}\text{Pu}$ per sample). As examples: daily urinary excretions of Pu in the general population are $\sim 10^6$ atoms;¹¹ plutonium bioassay programs require detection limits $\sim 10^6$ atoms per sample;¹⁴ mean plutonium concentrations in non-occupationally exposed persons are $\sim 10^6$ - 10^8 atoms per g of tissue;¹ and plutonium concentrations resulting from global fallout in the surface waters of the open ocean are $\sim 10^6$ - 10^8 atoms per L.^{4,21} Alpha spectrometry also does not provide $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic information, which is important for understanding systems with multiple sources. Depending on the source, $^{240}\text{Pu}/^{239}\text{Pu}$ ratios can vary from <1% to greater than 30%, and significant variations exist even within global fallout.⁵

A number of analytical methods were developed as sensitive alternatives to alpha spectrometry. These are fission track analysis (FTA),^{11,13,15,22-24} thermal ionization mass spectrometry (TIMS),^{4,5,7,14,25,26} inductively coupled plasma-mass spectrometry (ICP-MS),²⁷⁻³³

and resonance ionization mass spectrometry (RIMS).³⁴⁻³⁷ All of these techniques have demonstrated sensitivities of 10^6 - 10^8 atoms per sample, depending on the application and the level of interferences present, and all except FTA can provide $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratios.

Recently, accelerator mass spectrometry (AMS) has been demonstrated for the ultratrace analysis of plutonium concentrations and isotope ratios.^{38,39} AMS is a well-established method for the detection of long-lived radionuclides⁴⁰ and is an attractive alternative for plutonium measurements. AMS offers high efficiency, high rejection of interferences, low susceptibility to matrix components, and large dynamic range. These advantages may reduce demands on the sample preparation chemistry, which is a limiting factor for most other analytical methods. Ultimately AMS offers the potential combined benefits of high sample throughput and rapid analyses while maintaining low detection sensitivities of $\sim 1 \times 10^6$ atoms per sample for the plutonium isotopes $^{239,240,241,242,244}\text{Pu}$.

Capability for plutonium AMS measurements is under development at the Center for Accelerator Mass Spectrometry (CAMS)⁴¹ at Lawrence Livermore National Laboratory (LLNL).

2. AMS measurements of plutonium at LLNL

Heavy Element Beamline

Interference from uranium is a limiting factor in alpha spectrometry, FTA, and ICP-MS measurements of ^{239}Pu . Chemical separation procedures developed for these methods can produce quite low uranium backgrounds (10^8 - 10^{12} atoms per sample); however, these procedures are relatively expensive and laborious and typically dominate the effort and expense for analyses. Uranium also presents an interference for AMS, and the major technical issue for AMS measurements of Pu is rejection of ^{238}U by the spectrometer.⁴² Actinide development at CAMS is therefore focussed on reducing this background through abundance sensitivity. The installation of a Heavy Element spectrometer (see Figure 1) is central to this development.⁴² Improved abundance sensitivity, and the resulting high rejection of ^{238}U , provided by this spectrometer will

ease the requirements placed on the sample preparation chemistry while reducing background at mass 239 to below 1×10^6 atoms per sample.

A prototype of the Heavy Element spectrometer was used for the work reported here. Abundance sensitivity with this arrangement is provided by the low- and high-energy analyzing magnets. A Wien velocity filter provides additional rejection of some interferences, but cannot resolve ^{239}Pu ions from the small fraction of ^{238}U ions which pass through the analyzing magnets. The final spectrometer, presently under construction, includes a 45° cylindrical electrostatic analyzer (ESA) designed to remove the remaining ^{238}U .

Sample Preparation

Sample preparation is based on the standard protocols used for alpha spectrometry in the LLNL Marshall Islands Program,⁴³ with only minor modifications to provide a sample form appropriate for introduction into the ion source. A ^{242}Pu spike is added to samples prior to digestion for isotope dilution normalization and yield measurement.

For the IAEA samples reported below, five 10% splits of the processed digest (in HCl, following elution from the final column) were taken for AMS prior to electrodeposition onto alpha spectrometry planchets. AMS aliquots were combined with iron carrier (1 mg Fe as iron nitrate in commercial atomic absorption standard solution); then precipitated with iron hydroxide by addition of high purity ammonium hydroxide. Following rinses, the precipitates were re-dissolved in high purity nitric acid; transferred to small quartz crucibles; taken to dryness; and baked at 800°C . The resulting oxide was mixed with ~ 1 mg of high purity niobium powder, then pressed into a standard LLNL aluminum AMS sample holder.

AMS Measurement

A cesium sputter source⁴⁴ was used to produce 28 keV negative ions. The low energy spectrometer was sequentially cycled to accept the relevant analyte ions, $^{239-242}\text{Pu}^{16}\text{O}$ and $^{238}\text{U}^{16}\text{O}$. The high energy spectrometer was set to select 5+ ions at 39 MeV energy, and the high

energy analyzing magnet and Wien filter were cycled with the low energy spectrometer. At present, switching of the high energy magnet requires ~30 s, limiting precision and throughput. A gas ionization detector used for ion detection and particle identification allowed clean rejection of interfering ions at 4+ and lower charge states.

The AMS results reported below consisted of a single cycle through the masses (242, 238, 239, 240, 241, 242) for each sample. Measurement time was typically 30 s per isotope, and less than 10 min per sample. Count rates at each mass, corrected for electronics deadtime, were normalized to the average 242 rate for that sample, then multiplied by the known ^{242}Pu spike activity. No corrections for uranium or other backgrounds, or for mass bias, were employed.

3. Results

Total spectrometer rejection of ^{238}U (abundance sensitivity) with the prototype spectrometer is $\sim 1\text{--}4 \times 10^{-5}$. The ESA is expected to improve abundance sensitivity by a factor of 10-100, at which point abundance sensitivity is expected to be limited by other effects such as energy stability of the accelerator, $^{238}\text{U}^{16}\text{O}^+\text{H}^-$ and $^{238}\text{U}^{17}\text{O}^+$ ion formation, and scattering in the high energy spectrometer. Ultimately, overall sensitivity is expected to be limited by Pu introduced during sample preparation (process blank).

For soil samples and reagent blanks, backgrounds with the prototype spectrometer are consistently $\sim 1\text{--}2 \times 10^7$ atoms per sample at masses 237 and 239, limited by ^{238}U interference and possibly process blank. Backgrounds at masses 240 and 241 are $< 5 \times 10^6$ atoms per sample, limited by counting efficiency for 30 s measurements. No other significant interferences were observed at masses 237, 239, 240, or 241. Total efficiency (detected counts per Pu atom in the sample) is $> 2 \times 10^{-5}$ for 30 min measurements. Improvement by a factor of 2 or more can potentially be obtained with longer counting times and use of 4+ ions. Improvements in the resolution of the low energy spectrometer may also allow the ion source to be run at higher output, further increasing count rates.

Results for IAEA reference materials are shown in Tables 1 and 2. The results are compared to alpha spectrometry measurements, IAEA reference values, and to recent ICP-MS measurements reported by Muramatsu, *et al.*³² The $^{239+240}\text{Pu}$ activities as measured by AMS (Table 1) show a significant bias with increasing activity relative to the IAEA reference values (from +20% to -20%). However, the AMS results agree with the corresponding alpha spectrometry aliquots, indicating that the bias is not related to the AMS measurement.

The instrumental precision/accuracy is limited by time-dependence in the negative ion output. A fast switching mode and multiple passes for each sample will be used for routine measurements and will average out this time-dependence. Ultimately, a precision of 1-3% is expected.

The AMS results for $^{240}\text{Pu}/^{239}\text{Pu}$ isotope ratios in IAEA-135 showed good agreement to the published ICP-MS results. Agreement (within the AMS uncertainties) was also obtained for IAEA Soil 6. ^{241}Pu activities were also measured in the samples to examine our capabilities for other isotopes, and good agreement was observed for IAEA-367, the only sample for which we had an independent measurement.

3. Conclusions

The present work demonstrates that the CAMS AMS spectrometer is well-suited for providing high sensitivity, robust, high throughput measurements of plutonium concentrations and isotope ratios. Present backgrounds are $\sim 2 \times 10^7$ atoms per sample for environmental samples prepared using standard alpha spectrometry protocols. Measurements of Pu concentrations and isotope ratios in IAEA reference materials showed good agreement with reference values, alpha spectrometry, and ICP-MS results. Installation of the ESA is expected to reduce background levels below 1×10^6 atoms per sample at masses 237, 239, 240, and 241 while allowing simplifications of the sample preparation chemistry. Additional hardware and software improvements will allow measurements of plutonium concentrations and ratios for >50

unknowns in a 24 h period, which is typical for many of the long-lived radionuclides measured at CAMS.

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Figure Captions

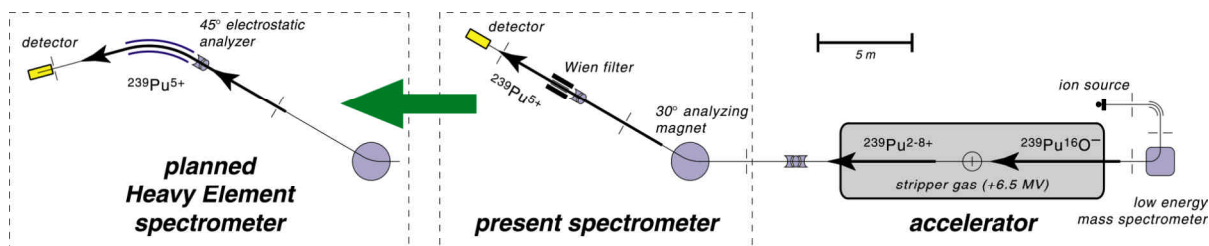


Figure 1 Accelerator mass spectrometry measurements of Pu isotopes at LLNL. Shown are the present setup, and planned upgrade (lower inset). In the present high energy spectrometer, a Wien filter is used to provide limited velocity analysis for the rejection of some interferences. In the final spectrometer, an electrostatic analyzer (cylindrical, 4.4 m radius, 5 cm plate gap, 45° bending angle, 50 kV/cm maximum field) will provide the final separation of interferences. The ESA has been designed to fully resolve neighboring isotopes at 250 AMU.

Table 1. Pu-239+240 activity concentrations. Recent measurements of ²³⁹Pu+²⁴⁰Pu activity concentrations (in Bq/kg) in IAEA reference materials by accelerator mass spectrometry (AMS) and alpha spectrometry at Lawrence Livermore National Laboratory. Two of five 10% aliquots (following sample digestion and chemical purification) were analyzed by AMS. One 50% aliquot was analyzed by alpha spectrometry. The results are compared to IAEA reference values.

IAEA Sample	IAEA reference (Bq/kg)	AMS result ¹ (Bq/kg)	α -spec result (Bq/kg)	AMS / IAEA %	α -spec / IAEA %	Sample Size ² (g)
IAEA-135 (Irish Sea Sediment)	213	244 \pm 2 246 \pm 2	252	115 % 115 %	118 %	1
IAEA-367 (Pacific Ocean Sed.)	38	36.6 \pm 0.2 31.9 \pm 0.2	38.5	96 % 84 %	101 %	10
IAEA, Soil 6 (Austrian Soil)	1.04	0.96 \pm 0.04 0.92 \pm 0.04	0.97	92 % 88 %	94 %	10
IAEA-375 (Chernobyl Soil)	0.30	0.27 \pm 0.02 0.21 \pm 0.02	0.23	91 % 70 %	77 %	10
Reagent Blank (10 g equivalent)	—	0.014 \pm 0.006 0.014 \pm 0.007	<0.1	— —	— —	— —

¹ The AMS results have not been corrected for U-238 interference. Quoted AMS uncertainties include counting statistics only.

² Total sample mass digested. Five 10% aliquots of the digest were collected for AMS measurements, and the remaining was analyzed by alpha-spectrometry.

Table 2. Pu-240/239 isotope ratios and Pu-241 activity concentrations.
Recent measurements of plutonium isotopes in IAEA reference materials by accelerator mass spectrometry (AMS) and alpha spectrometry at Lawrence Livermore National Laboratory. The results are compared to recently published results of Muramatsu, *et al.*, measured by ICP-MS.³²

IAEA Sample	240/239 ICP-MS result ³ %	240/239 AMS result ⁴ %	241 activity AMS result ^{4,5} (Bq/kg)
IAEA-135 (Irish Sea Sediment)	21.1 ± 0.4	21.3 ± 0.6 21.1 ± 0.6	2910 ± 180 2940 ± 200
IAEA, Soil 6 (Austrian Soil)	19.1 ± 0.5	17 ± 4 18 ± 4	1 ± 2 6 ± 3
IAEA-367 (Pacific Ocean Sediment)	—	32.0 ± 0.6 29.6 ± 0.5	116 ± 10 ⁶ 90 ± 8 ⁶

³ ICP-MS results reported by Muramatsu, *et al.* (Ref. 32).

⁴ The AMS results have not been corrected for U-238 interference. Quoted AMS uncertainties include counting statistics only.

⁵ Pu-241 activity on 8-AUG-1999.

⁶ The IAEA provides an information value for Pu-241 activity in this sample of 170 Bq/kg referenced to 1-JAN-1990 (see Ref. 45). Decayed to 8-AUG-1999 this information value becomes 107 Bq/kg.

Bibliography

- 1 D. M. Taylor, *Environmental Plutonium in Humans*, Appl Radiat Isotopes **46**, 1245-1252 (1995).
- 2 R. J. Pentreath, *The Analysis of Pu in Environmental Samples - a Brief Historical Perspective*, Appl Radiat Isotopes **46**, 1279-1285 (1995).
- 3 R. J. Cornett, T. Eve, A. E. Docherty, and E. L. Cooper, *Plutonium in Freshwaters - Sources and Behaviour in the Ottawa River Basin*, Appl Radiat Isotopes **46**, 1239-1243 (1995).
- 4 P. J. Kershaw, K. E. Sampson, W. McCarthy, and R. D. Scott, *The Measurement of the Isotopic Composition of Plutonium in an Irish Sea Sediment By Mass Spectrometry*, J Radioanal Nucl Chem Art **198**, 113-124 (1995).
- 5 K. O. Buesseler, *The isotopic signature of fallout plutonium in the North Pacific*, J Environ Radioact **36**, 69-83 (1997).
- 6 P. J. Kershaw, D. C. Denoon, and D. S. Woodhead, *Observations on the redistribution of plutonium and americium in the Irish Sea sediments, 1978 to 1996: concentrations and inventories*, J Environ Radioact **44**, 191-221 (1999).
- 7 A. B. Kersting, D. W. Efurdu, D. L. Finnegan, D. J. Rokop, D. K. Smith, and J. L. Thompson, *Migration of plutonium in ground water at the Nevada Test Site*, Nature **397**, 56-59 (1999).
- 8 L. C. Sun, C. B. Meinhold, A. R. Moorthy, E. Kaplan, and J. W. Baum, *Assessment of plutonium exposure in the Enewetak population by urinalysis*, Health Phys **73**, 127-132. (1997).
- 9 R. A. Guilmette, *Issues and research on the biochemistry of inhaled actinides*, J Alloys Compounds **271**, 66-71 (1998).
- 10 J. C. Nenot and J. W. Stather, *The toxicity of plutonium, americium and curium* (Pergamon Press, London, 1979).
- 11 M. E. Wrenn, N. P. Singh, and Y. H. Xue, *Urinary Excretion of Pu-239 By the General Population - Measurement Technique and Results*, Radiat Prot Dosim **53**, 81-84 (1994).
- 12 B. Franke, R. Schupfner, H. Schuttelkopf, and D. H. R. Spennemann, *Transuranics in Bone of Deceased Former Residents of Rongelap Atoll, Marshall Islands*, Appl Radiat Isotopes **46**, 1253-1258 (1995).
- 13 L. C. Sun, A. R. Moorthy, E. Kaplan, J. W. Baum, and C. B. Meinhold, *Assessment of Plutonium Exposures in Rongelap and Utirik Populations By Fission Track Analysis of Urine*, Appl Radiat Isotopes **46**, 1259-1269 (1995).
- 14 W. C. Inkret, D. W. Efurdu, G. Miller, D. J. Rokop, and T. M. Benjamin, *Applications of thermal ionization mass spectrometry to the detection of Pu-239 and Pu-240 intakes*, Int J Mass Spectrom **178**, 113-120 (1998).
- 15 M. P. Krahenbuhl and D. M. Slaughter, *Improving process methodology for measuring plutonium burden in human urine using fission track analysis*, J Radioanal Nucl Chem **230**, 153-160 (1998).
- 16 J. H. Shinn, D. N. Homan, and W. L. Robison, *Resuspension studies in the Marshall Islands*, Health Phys **73**, 248-257. (1997).
- 17 D. L. Donohue, *Strengthening IAEA safeguards through environmental sampling and analysis*, J Alloys Compounds **271**, 11-18 (1998).
- 18 B. Salbu, T. Krekling, and D. H. Oughton, *Characterisation of radioactive particles in the environment*, Analyst **123**, 843-849 (1998).
- 19 W. Templeton, F. Harrison, J. Knezovich, N. Fisher, and D. Layton, *Bioconcentration of Radionuclides in Marine Food-Web Organisms in Radionuclides in the Arctic Seas from the Former Soviet Union: Potential Health and Ecological Risks*, edited by D. Layton, L. C. D. R. R. Edson, M. Varela, and B. Napier (Arctic Nuclear Waste Assessment Program (ANWAP), Office of Naval Research (ONR), 1997), p. 4-1/4-12.
- 20 P. K. Kuroda, *Extraterrestrial Radioactivity*, J Radioanal Nucl Chem Art **203**, 591-599 (1996).
- 21 V. T. Bowen, K. M. Wong, and V. E. Noshkin, *Plutonium-239 in and over the Atlantic Ocean*, J Mar Res **29**, 1-0 (1971).

- 22 A. Moorthy, C. J. Schopfer, and S. Banerjee, *Plutonium from atmospheric weapons testing: fission rack analysis of urine samples.*, Anal Chem **60**, 857A (1988).
- 23 L. Johansson and E. Holm, *Determination of Trace-Amounts Pu-239 Using Fission Track Analysis*, Nucl Instrum Meth Phys Res a **376**, 242-247 (1996).
- 24 L. Johansson, C. Samuelsson, and E. Holm, *Evaluation of the fission track analysis for determination of trace-amounts of Pu-239*, Nucl Instrum Meth Phys Res a **423**, 453-460 (1999).
- 25 K. O. Buesseler and J. E. Halverson, *The mass spectrometric determination of fallout 239Pu and 240Pu in marine samples.*, J Envir Rad **5**, 425 (1987).
- 26 K. O. Buesseler, S. A. Casso, M. C. Hartman, and H. D. Livingston, *Determination of Fission-Products and Actinides in the Black Sea Following the Chernobyl Accident*, J Radioanal Nucl Chem Art **138**, 33-47 (1990).
- 27 J. S. Crain and J. Alvarado, *Hydride Interference On the Determination of Minor Actinide Isotopes By Inductively Coupled Plasma Mass Spectrometry*, J Anal Atom Spectrom **9**, 1223-1227 (1994).
- 28 R. Chiappini, J. M. Taillade, and S. Brebion, *Development of a High-Sensitivity Inductively Coupled Plasma Mass Spectrometer For Actinide Measurement in the Femtogram Range*, J Anal Atom Spectrom **11**, 497-503 (1996).
- 29 J. M. B. Moreno, M. Betti, and J. I. G. Alonso, *Determination of neptunium and plutonium in the presence of high concentrations of uranium by ion chromatography inductively coupled plasma mass spectrometry*, J Anal Atom Spectrom **12**, 355-361 (1997).
- 30 A. E. Eroglu, C. W. McLeod, K. S. Leonard, and D. McCubbin, *Determination of plutonium in seawater using co-precipitation and inductively coupled plasma mass spectrometry with ultrasonic nebulisation*, Spectrochim Acta Pt B-At Spec **53**, 1221-1233 (1998).
- 31 J. S. Becker, H. J. Dietze, J. A. McLean, and A. Montaser, *Ultratrace and isotope analysis of long-lived radionuclides by inductively coupled plasma quadrupole mass spectrometry using a direct injection high efficiency nebulizer*, Anal Chem **71**, 3077-3084 (1999).
- 32 Y. Muramatsu, S. Uchida, K. Tagami, S. Yoshida, and T. Fujikawa, *Determination of plutonium concentration and its isotopic ratio in environmental materials by ICP-MS after separation using ion-exchange and extraction chromatography*, J Anal Atom Spectrom **14**, 859-865 (1999).
- 33 I. Rodushkin, P. Lindahl, E. Holm, and P. Roos, *Determination of plutonium concentrations and isotope ratios in environmental samples with a double-focusing sector field ICP-MS*, Nucl. Instrum. Methods Phys. Res. A, Accel. Spectrom. Detect. Assoc. Equip. (Netherlands) **423**, 472-9 (1999).
- 34 G. Passler, N. Erdmann, H. U. Hasse, G. Herrmann, G. Huber, S. Kohler, J. V. Kratz, A. Mansel, M. Nunnemann, N. Trautmann, and A. Waldek, *Application of laser mass spectrometry for trace analysis of plutonium and technetium*, Kerntechnik **62**, 85-90 (1997).
- 35 B. Eichler, S. Hubener, N. Erdmann, K. Eberhardt, H. Funk, G. Herrmann, S. Kohler, N. Trautmann, G. Passler, and F. J. Urban, *An atomic beam source for actinide elements: Concept and realization*, Radiochim Acta **79**, 221-233 (1997).
- 36 M. Nunnemann, N. Erdmann, H. U. Hasse, G. Huber, J. V. Kratz, P. Kunz, A. Mansel, G. Passler, O. Stetzer, N. Trautmann, and A. Waldek, *Trace analysis of plutonium in environmental samples by resonance ionization mass spectroscopy (RIMS)*, J Alloys Compounds **271**, 45-48 (1998).
- 37 K. Wendt, K. Blaum, B. A. Bushaw, C. Gruning, R. Horn, G. Huber, J. V. Kratz, P. Kunz, P. Muller, W. Nortershauser, M. Nunnemann, G. Passler, A. Schmitt, N. Trautmann, and A. Waldek, *Recent developments in and applications of resonance ionization mass spectrometry*, Fresenius J Anal Chem **364**, 471-477 (1999).
- 38 L. K. Fifield, R. G. Cresswell, M. L. Ditada, T. R. Ophel, J. P. Day, A. P. Clacher, S. J. King, and N. D. Priest, *Accelerator Mass Spectrometry of Plutonium Isotopes*, Nucl Instrum Meth Phys Res B **117**, 295-303 (1996).
- 39 L. K. Fifield, A. P. Clacher, K. Morris, S. J. King, R. G. Cresswell, J. P. Day, and F. R. Livens, *Accelerator mass spectrometry of the planetary elements*, Nucl Instrum Meth Phys Res B **123**, 400-404 (1997).
- 40 J. S. Vogel, K. W. Turteltaub, R. Finkel, and D. E. Nelson, *Accelerator Mass Spectrometry - Isotope Quantification At Attomole Sensitivity*, Anal Chem **67**, A353-A359 (1995).

- 41 M. L. Roberts, G. S. Bench, T. A. Brown, M. W. Caffee, R. C. Finkel, S. Freeman, L. J. Hainsworth, M. Kashgarian, J. E. McAninch, I. D. Proctor, J. R. Southon, and J. S. Vogel, *The LLNL AMS facility*, Nucl Instrum Meth Phys Res B **123**, 57-61 (1997).
- 42 J. E. McAninch and T. F. Hamilton, *Measurement of plutonium and other actinides at the Center for Accelerator Mass Spectrometry: A comparative assessment of competing techniques*, Lawrence Livermore National Laboratory, Report No. UCRL-ID-133118 (1999).
- 43 K. M. Wong, T. Jokela, and V. E. Noshkin, *Radiochemical procedures for analysis of Pu, Am, Cs, and Sr in water, soil, sediments and biota samples*, Lawrence Livermore National Laboratory, Report No. UCRL-ID-116497 (1994).
- 44 J. R. Southon and M. L. Roberts, *Ten years of sourcery at CAMS/LLNL — evolution of a Cs ion source, These proceedings.* (1999).